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16. ABSTRACT

Disinfection of drinking water has been used for many years for the prevention of waterborne disease through inactivation of pathogenic microorganisms. In general, disinfection practices in the field have developed through experience rather than application of specific disinfection data derived from controlled experiments. Concerns about waterborne disease outbreaks caused by the protozoan Giardia lamblia, the cysts of which are very resistant to inactivation, and the need to more precisely control the disinfection process to reduce formation of potentially hazardous chemical byproducts have made it necessary to more closely examine the biocidal aspects of drinking water disinfection. The use of mathematical concepts based on microbial inactivation kinetics to predict disinfectant concentration and contact times needed to inactivate specific proportions of various pathogens and indicator species has received increasing attention as an approach to better control of the disinfection process. This report describes the use of these concepts, the problems in extrapolation from the available data and in application of laboratory disinfection data to field conditions.

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INTRODUCTION

The primary purpose of the disinfection process in drinking water treatment is the control of waterborne diseases through inactivation of any pathogenic microorganisms present in the water. The high degree of success of this process used alone or in combination with other treatment processes is confirmed by the infrequent occurrence of waterborne disease outbreaks in the U.S. in modern times.

Interestingly, the disinfectant concentrations and contact times used by different water utilities vary widely. There are no Federal requirements regarding these parameters, but several states and advisory groups provide minimum requirements or recommendations. For instance, Washington State regulations require a free-chlorine residual of 0.2 mg/L after 30 min contact. Other recommended standards (1) call for 0.2 to 0.5 mg/L free residual chlorine with 30 min contact for groundwaters or 120 min contact for surface waters. For chloramines, 1 to 2 mg/L with 120 and 180 min contact time for groundwaters and surface waters, respectively, is recommended. Regulations and recomendations such as these have been developed and formalized over a period of years and are based in part on disinfection data and in part on practices that seemed prudent and that have consistently produced finished water that would meet coliform indicator standards.

Interest in more soundly based approaches to establishment of disinfection requirements for drinking water treatment has been stimulated primarily by two factors: 1) the recognition that the cysts of Giardia lamblia, an important cause of waterborne disease outbreaks, are extremely resistant to inactivation by disinfectants and 2) the reliance on disinfection only as an accepted drinking water treatment practice. This practice is especially prevalent in systems using high quality surface source waters relatively free from human fecal contamination but subject to Giardia cyst contamination from other animal sources.

Currently, a more rigorous approach, based on disinfectant concentration (C) and contact time (t) or C·t values required to inactivate different types of pathogens to certain levels under specified conditions as established by laboratory experimentation, has received considerable attention. In this report, the basis for use of this concept and some of the advantages and disadvantages of its application to drinking water disinfection will be discussed.

DETERMINATION OF C·t VALUES

C·t values are determined according to the following equation:

$$k = C \cdot t$$

in which C is the concentration of a specific disinfectant (mg/L)

t is the contact time (min) required for inactivation of a specific proportion of the microorganism (e.g. 99%)

k is a constant for a specific microorganism exposed under specific conditions

Thus, C·t values can be defined as the product of the disinfectant concentration (C) and contact time (t) required for inactivation of a defined proportion of a specific pathogen by a specific disinfectant under specified temperature and pH conditions.

ORIGIN OF C·t CONCEPT, WATSON'S LAW

The C·t concept is based on an empirical equation developed by Watson (2) to examine the effects of changing disinfectant concentration on rates of microbial inactivation. Watson's Law is:

$$k = C^{n} \cdot t$$

The terms are the same as in the k = C·t equation except for an important additional term n which is a constant also called the "coefficient of dilution." Watson's Law is similar to the van't Hoff equation (3, 4) used for determining the nature of chemical reactions in which the value n determines the order of the chemical reaction.

In applying this equation to disinfection studies, the results of a number of individual disinfection experiments using different disinfectant concentrations are used. The disinfectant concentrations (C) and times (t) necessary to attain a specific degree of inactivation (e.g. 99%) are plotted on double logarithmic paper. Such plots result in a straight line with slope n (5, 6). Fig. 1 illustrates data plotted in this manner and also the significance of the value of n in extrapolation of disinfection data. When n = 1, the C·t value remains constant regardless of the disinfectant concentration used. When n = 2, C·t values decline as disinfectant concentrations increase while when n = 0.5, C·t values increase as disinfectant concentrations increase. Thus, when n = 1, disinfectant concentration and exposure time are of equal importance in determining the inactivation rate, or the C·t product, k. If n > 1, disinfectant concentration is the dominant factor in determining the inactivation rate while if n < 1, exposure time is more important than disinfectant concentration.

Thus, the value of n is a very important factor in determining the degree to which extrapolation of data from disinfection experiments is valid. If n = 1, C·t values can be used to predict disinfection efficiency over a broad range of disinfectant concentrations and contact times. Haas and Karra (7) calculated n values from a number of older free and combined chlorine disinfection studies conducted under disinfectant demand-free conditions. The calculated

values are summarized in Table 1. If one accepts n values in the range 0.7 to 1.3 as being within the range of experimental error from 1.0, most of the values can be considered as representing 1. Few n values were < 0.7 but a considerable number were > 1.3. Additional n values reported in more recent studies and for disinfectants other than chlorine are shown in Table 2. With few exceptions, the values are near 1. One value, 0.34 for inactivaton of G. muris at pH 7, 5°C in the free chlorine range of 0.41 to 2.73 mg/L illustrates the significance of n values. This value indicates that C·t values increase substantially as chlorine concentration is increased in the range. Other data from the study cited indicate that C·t values for 99% inactivation of G. muris cysts under these conditions (pH 7, 5°C) increase from 153 at 0.41 mg/L to 630 at 2.73 mg/L.

APPLICATION OF THE C·t CONCEPT TO DISINFECTION PRACTICE

Baumann and Ludwig (8) applied Watson's Law ($k = C^n \cdot t$) in illustrating a procedure that could be used in making disinfection recommendations for small nonpublic water supplies that relied on disinfection only for pathogen control. Using data from disinfection studies by others they developed double logarithmic plots of C vs. t for two different temperature ranges, 20-29°C and 0-5°C. They based their C·t value recommendations on constant C·t values that included certain pathogens of concern and desired safety factors. The n values based on experimental data for several pathogens deviated from 1. They commented on the apparent inadequacy of the coliform group, exemplified by E. coli, to indicate adequate disinfection requirements for viral agents and protozoans. In plotting the curves, however, they used data from different studies representing from 99.6 to 100% inactivation. Therefore, the curves for different microorganisms are not directly comparable.

Following this, the C·t value concept received little further attention until its use in a comprehensive review of the disinfection literature by the Safe Drinking Water Committee (9). Although the Committee chose C·t values as the method for comparing biocidal efficacy, they did not delve into the background of its development nor did they attempt extrapolation to other values for either C or t from those calculated from available data. Following publication of the Committee report, the use of C·t values for interpreting disinfection data became more prevalent, both for assessing comparative efficacies of different disinfectants and for expressing comparative resistances of different types of pathogens (bacteria, viruses, cysts). Wickramanayake et al. (10), studying the inactivation of Giardia and Naeglaria cysts by ozone, concluded that:

> "Within the reported ranges of temperatures, pH, and ozone concentrations, the cyst inactivation kinetics deviated from Chick's law because there was an initial lag period. The rate of inactivation increased during this lag period and then declined where the inactivation rate was directly proportional to the number of viable cysts. At 99% inactivation, the results were well represented by Watson's law with coefficient of dilution being close to one. Consequently, the empirical constant k of Watson's law or the C·t product can be used with confidence to compare the effectiveness of disinfectants or the sensitivity of cysts."

It is important to note that Wickramanayake et al. (10) specified that C·t values could be used to compare disinfectant effectiveness at the 99% inactivation level. The 99% inactivation level has been used in most studies for calculating C·t values. It is likely that this level of inactivation was originally selected by the Safe Drinking Water Committee (9) because it is at this level that exponential kinetics are usually best approximated. Ideally, if exponential kinetics were followed and if C·t values for 99% inactivation were known one could easily calculate C·t values for other degrees of inactivation. The problems associated with the validity of such calculated C·t values are discussed in the next section.

DISINFECTION KINETICS

As indicated, C·t values are usually based on the disinfectant concentration and contact time needed for 99% inactivation of various target microorganisms. The validity of extrapolations from C·t values required for 99% inactivation to C·t values required for other levels of inactivation (50%, 90%, 99.9%) is dependent on the nature of the inactivation curves from which the 99% inactivation C·t value was determined.

Chick (11) characterized the inactivation of microorganisms as analogous to a first order chemical reaction with the disinfectant and the microorganisms constituting the reactants. This analogy is generally expressed as Chick's Law which may be written as follows:

$$\log {}^{N}/_{N_0} = - K t$$

where ${}^{N}/_{N_0}$ is the fraction of the original number of organisms remaining at time, t, and K is a proportionality constant. Ideally, plots of log ${}^{N}/_{N_0}$ against t for various contact times should provide a straight line (See Fig. 2). In actual experiments, deviations from first order kinetics are common and can be attributed to several causes. A major factor is the failure of microorganisms to behave as chemical reactants. Microorganisms are much more complex than simple chemical reactants and many different chemical reactions may be involved in their inactivation. For the measured activity, inactivation, to occur, diffusion of the disinfectant into the microorganism and reaction with one or more vital systems is necessary. In addition, microbial populations are much less physically and chemically homogenous than chemical reactants, further contributing to variations in inactivation rates. Aggregation or clumping of a portion of the population or association with extraneous material may delay disinfectant action. Experimental conditions can also influence the shape of the inactivation curve. These include the order in which the reactants (disinfectant and microorganism) are added to the reaction vessel, mixing efficiency, and depletion of disinfectant concentration through extraneous reactions (disinfectant demand) or volatilization of the disinfectant.

Some of the types of curves generated in disinfection experiments are shown in Fig. 3 (12). Fig. 3 A demonstrates true exponential or first order kinetics, a straight line passing through the zero time, 100% survival point. Fig. 3 B shows concave upward kinetics or an initial "shoulder" followed by exponential kinetics. Curves of this type are usually encountered with more resistant types

of microorganisms or weak biocides, perhaps caused by delays in diffusion of the disinfectant to sites of action. The initial lag periods can also be anomalies caused by inadequate mixing. Fig. 3 C shows concave downward kinetics, an initial rapid decline, followed by a slower exponential rate. Possible causes include differential sensitivity of portions of the microbial population or disinfectant depletion. Fig. 3 D shows multiple kinetics or a "tailing off" curve. This type of curve may be caused by presence of an innately more resistant fraction of the population, protection of a portion of the population by extraneous material, or disinfectant depletion. Other combinations of the illustrated curves also may occur. It should be emphasized that failure of the results of disinfection experiments, including most of Chick's original studies, to result in the exponential curve predicted by Chick's Law is the normal occurrence rather than the exception (6).

The failure of microbial inactivation to conform to first order kinetics has important implications relative to the use of C·t values for predicting microbial inactivation. If first order kinetics were followed, it would be simple to calculate C·t values for other levels of inactivation (e.g. inactivation to the 99.99% level would require a C·t value twice that required for 99% inactivation. Deviations of the "tailing off" type frequently seen are particularly troublesome because the calculated C·t value would be less than that actually required.

TEMPERATURE EFFECTS ON INACTIVATION RATES

C·t values are determined experimentally at specific temperature (e.g. 5°C, 15°C, 25°C) or often at only one temperature. For applications to field conditions it is necessary to understand the effects of water temperature changes on disinfection rates and C·t values.

The effects of changes in temperature on both chemical reaction rates and the biocidal activity of chemical disinfectants are well documented. Chemical reaction rates generally increase with increasing temperatures and, according to the empirical "common rule", rates increase by a factor of 2 to 3 for each 10°C rise in temperature (13). Disinfection rates also increase with increasing temperature. The coefficient by which disinfection rates increase for each 10°C rise in temperature is referred to as the Q_{10} value. Clarke and Chang (14) in reviewing temperature effects on inactivation rates observed that Q_{10}'s for viral destruction were in the range of 2 to 3. Q_{10}'s calculated from a number of disinfection studies involving different biological agents and different disinfectants are shown in Table 3. The Q_{10} coefficients range from 1.42 to 5.17 with most falling in the range of 2 to 3. In three cases Q_{10} coefficients for temperature changes in the ranges 5°C to 15°C and 15°C to 25°C show large differences. In two cases 5°C to 15°C Q_{10}'s exceed those for 15°C to 25°C and in the other case, the reverse is true.

Some investigators have expressed a concern regarding the possibility that, as temperatures approach 0°C, disinfection rates may decrease by a larger factor than would be predicted by Q_{10} values in the ranges of 5°C to 15°C or 15°C to 25°C (15); (C. P. Hibler, Colorado State University, personal communication). Based on the physical laws that govern chemical diffusion and reaction rates in aqueous media, there is no basis for assuming any special retardation

of rates as temperatures approach 0°C. Few studies have been conducted at temperatures below 5°C and the results of those available are insufficient to be conclusive. In general, the "common rule" of a 2 to 3 fold increase in inactivation rates per 10°C increase in temperature seems fairly well substantiated.

EFFECTS OF DISINFECTANT CHARACTERISTICS

The disinfectants of interest, free chlorine, chloramines, chlorine dioxide, and ozone have individual characteristics that influence both the results of laboratory disinfection studies and their performance in the field.

Free Chlorine

Free chlorine exists in aqueous solution in two different forms, HOCl and OCl^-, the proportions of which vary in a nonlinear fashion according to pH. In addition, the HOCl form is usually estimated to be 150 to 300 fold more effective biocidally than OCl^-. Over a pH range of 6.0 to 10.0 these species are present in proportions of over 98% HOCl at pH 6.0 to over 99% OCl^- at pH 10.0. Between pH 7.0 and 8.5 the proportions change rapidly but nonlinearly, with HOCl decreasing from about 83% at pH 7.0 to about 14% at pH 8.5. The equilibrium is somewhat temperature dependent, with lower temperatures resulting in slightly increased proportions of HOCl. As indicated, the two chemical species are in equilibrium and as one species is consumed in reactions, the proportions remain the same, the concentration of each being decreased proportionally. Chlorine losses caused by volatilization normally are not a problem in experiments but decreases in concentration caused by reactions with chlorine demanding substances can be substantial.

Chloramines

Chloramines are the most chemically complex of the four disinfectants. Although combined chlorine exists in a number of forms (NH_2Cl, $NHCl_2$, NCl_3) only the monochloramine form (NH_2Cl) is of major interest since it is the predominant species usually found under drinking water treatment conditions. Conditions that lead to formation of $NHCl_2$ and NCl_3 are avoided because they contribute to taste and odor problems (16). When chlorine and ammonia are mixed in equimolar proportions (weight ratios of chlorine to ammonia of 5:1) nearly all of the free chlorine is converted to monochloramine although this is an equilibrium reaction and a very small proportion of free chlorine remains. This equilibrium is affected by the chlorine to ammonia ratio. The rate of monochloramine formation is also pH dependent. It is formed most rapidly at pH 8.3, the rate of formation decreasing as pH decreases or increases from pH 8.3. Biocidally, monochloramine is considered to be a very weak disinfectant, requiring for equivalent inactivation contact times about 25 to 100 fold higher than for equal concentrations of free chlorine. The inactivation efficiency of monochloramine also is pH dependent with inactivation rates increasing as pH decreases. This has been attributed by some to the disinfectant activity of the free chlorine species present in equilibrium with monochloramine. At low pH's it would be predominantly in the form of the more effective free chlorine species, HOCl. Chloramine losses caused by volatilization usually are not a problem in experiments. Levels are usually quite stable

but unrecognized combination with certain organic compounds to form less biocidally efficient organic chloramines may occur (17, 18) and remain unrecognized because the tests for combined chlorine do not distinguish between inorganic and organic chloramines.

Application of laboratory experimental results to field conditions is especially difficult because laboratory studies have conventionally employed preformed chloramines, while in practice, chloramines are not preformed and varying degrees of inactivation may actually be caused by the transient presence of free chlorine prior to its combining with ammonia. This is further complicated by the fact that the rate of conversion to chloramine is pH dependent. Also, in laboratory studies chlorine to ammonia ratios of 1:2 to 1:6 often are used to minimize the presence of free chlorine while in field applications chlorine to ammonia ratios ranging from 2:1 to 5:1 are commonly used, thus influencing the free chlorine to combined chlorine equilibrium. Because of these complicating factors, Ward et al. (19) concluded that:

> "The effectiveness of chloramine compounds as primary disinfectants for drinking water can only be determined when the method of application, pH, and concentrations of chlorine and ammonia are considered."

It should be noted that the results of laboratory studies using preformed chloramines will always provide an underestimate of their effectiveness relative to field application. The differences can be substantial and the actual field relevance of C·t values based on such laboratory studies is questionable.

Chlorine Dioxide

Chlorine dioxide is less subject to disproportionation reactions than chlorine and chloramines within the pH ranges generally encountered in water treatment. At high pH's (above 10) chlorine dioxide disproportionates at increasing rates to chlorate and chlorite species that have no disinfecting capability (20). In contrast to free chlorine and chloramines, the disinfection efficiency of chlorine dioxide increases by a factor of about 3 fold as pH increases over a range of pH 6 to pH 9 or higher. Chlorine dioxide exists as an undissociated gas dissolved in water and at these pH's does not disproportionate into other chemical species. Therefore, the increased activity is caused by some other factor. Because it is present as an undissociated dissolved gas, chlorine dioxide is more easily lost through volatilization than free or combined chlorine. This may affect the kinetics in experiments with longer exposure times, particularly those conducted at higher temperatures. Chlorine dioxide is less likely than free or combined chlorine to react with disinfectant demand substances and therefore is more stable than chlorine in this regard both in laboratory experiments and under field conditions.

Ozone

Ozone is the most unstable and highly reactive of the four disinfectants. Like chlorine dioxide, ozone is present in water as a dissolved gas. Therefore, it is subject to losses through both volatilization and reactions with organic chemicals present in water. The results of most studies indicate that ozone biocidal efficiency

is relatively unaffected by pH within the range encountered in drinking water treatment (9). Because of its volatility and high reactivity, maintaining stable ozone levels either in laboratory experiments or under field conditions is very difficult. These factors also make C·t values for ozone less precise than for the other disinfectants.

C·t VALUES FOR VARIOUS PATHOGENS

C·t values for 99% inactivation at 5°C of a variety of pathogens by free and combined chlorine, chlorine dioxide, and ozone are shown in Table 4. It is evident that Giardia cysts are the most resistant of all the microorganisms shown to all of the disinfectants except combined chlorine. Viruses are more resistant than cysts to this disinfectant. Ozone is the most effective disinfectant overall. Free chlorine and chlorine dioxide are approximately equivalent at the pH's indicated except that chlorine dioxide is considerably more effective than free chlorine for cyst inactivation.

CONCLUSIONS

The major advantage of the use of C·t values for establishing disinfectant concentrations and contact times needed for disinfection of drinking water is that the values are based on experimental data. This is particularly important in situations in which disinfection is the only treatment process used. In the absence of treatment processes that physically remove microorganisms, disinfection must be able to inactivate any pathogens present in the source water to a sufficient degree to prevent waterborne disease.

A major problem associated with the use of C·t values is that because the experimental data base is extremely limited, one must extrapolate from C·t values determined for a limited number of experimental conditions to the wide range of water temperatures and pH's experienced under field conditions. In some cases essential data are not available. While the effects of temperature changes on disinfection rates seem to be fairly predictable, pH effects are difficult to take into account, particularly for free and combined chlorine. That disinfectant concentration and exposure time are equally important in determining disinfectant effectiveness seems substantiated in many cases but some data do not support this.

Extrapolation from 99% inactivation levels to other levels is also questionable because of the frequent failure of inactivation rates to follow Chick's Law. It is likely that rates would be even less predictable under field conditions because of the state of the microorganisms and the possibility of protective effects.

In view of these problems and questions, the use of C·t values to establish disinfection requirements for drinking water treatment must be approached cautiously and conservatively.

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Table 1. Values of $\underline{n}$ for inactivation of microorganisms by chlorine[a,b]

Disinfectant	Number of Studies	$\underline{n}$ Value Range	Number of $\underline{n}$ Values		
			< 0.7	$0.7-1.3$	> 1.3
Free chlorine	34	0.6-7.3	2	23	9
Combined Chlorine	39	0.5-13.3	1	22	16

[a] $\underline{n}$ = The exponent of C in the Watson equation ($k = C^{n}$).
[b] $\underline{n}$ values calculated by Haas and Karra (7) using data from various sources.

Table 2. Additional values of $\bar{n}$ for free chlorine, ozone, and chlorine dioxide

Disinfectant	Organism	pH	Temp. (C)	Range Disinfect. Conc. (mg/L)	$\bar{n}$	Reference
Free Chlorine	*N. gruberi*	5.0	25	0.49-2.68	0.96	(21)
		7.0	25	0.78-3.44	1.19	
		9.0	25	11.6-72.6	0.93	
Free Chlorine	*G. muris*	7.0	5	0.41-2.73	0.34	(22)
		7.0	5	11.1-78.5	1.52	
		7.0	5	186-244	4.76	
		5.0	25	4.9-13.0	1.35	
		7.0	25	2.87-7.12	1.59	
		9.0	25	15.5-84.1	0.90	
Chlorine Dioxide	polio 1	7.0	5	0.4-1.0	1.05	(23)
		7.0	25	0.4-0.8	1.02	
Chlorine Dioxide	*E. coli*	6.5	5	0.25-0.75	1.08	(24)
		6.5	10	0.25-0.75	1.18	
		6.5	20	0.25-0.75	1.08	
Chlorine Dioxide	*G. muris*	7.0	5	0.10-5.55	1.20	(22)
		5.0	25	0.26-1.20	1.22	
		7.0	25	0.26-1.12	1.30	
		9.0	25	0.15-0.81	1.37	
Ozone	polio 1	7.2	20	0.1-0.21	3.4	(25)
Ozone	*N. gruberi*	5-9	25	0.21-1.05	1.0	(10)
		7	5-30	0.3-2.0	1.1	
Ozone	*G. muris*	5-9	25	0.02-0.19	1.2	(10)
		7	5-25	0.03-0.7	1.1	

Table 3. Effects of 10°C changes in temperature (Q_{10}) on microorganism inactivation rates

Disinfectant	Organisms	Temp. (C)	pH	Q_{10}	Reference
Free chlorine	*E. coli*	5-25	7	1.65	(6)
			8.5	1.42	
			9.8	2.13	
			10.7	2.5	
	E. histolytica cysts	-	-	2.1	
Chloramines	*E. coli*	5-25	7.0	2.09	(6)
			8.5	2.28	
			9.5	3.35	
	Polio 1	5-15	9.0	1.5	(26)
		15-25	9.0	4.0	(99% inactivation)
	"	5-15	9.0	2.0	(26)
		15-25	9.0	1.9	(99% inactivation)
	"	5-15	4.5	2.5	
ClO_2	Polio 1	5-15	7	2.26	(26)
		15-25	7	1.99	
		5-15	7	4.12	(27)
		15-25	7	1.34	
O_3	Polio 1	10-20		1.5	(25)
	N. gruberi	5-15	7	2.07	
	"	15-25	7	1.58	(10)
	G. muris	5-15	7	5.17	
		15-25	7	1.39	

Table 4. Summary of C·t value ranges for 99% inactivation of various microorganisms by disinfectants at 5°C

Micro-organism	Disinfectant			
	Free Chlorine pH 6 to 7	Preformed Chloramine pH 8 to 9	Chlorine Dioxide pH 6 to 7	Ozone pH 6 to 7
E. coli	0.034-0.05	95-180	0.4-0.75	0.02
Polio 1	1.1-2.5	768-3740	0.2-6.7	0.1-0.2
Rotavirus	0.01-0.05	3806-6476	0.2-2.1	0.006-0.06
Phage f_2	0.08-0.18	-	-	-
G. lamblia cysts	47->150	-	-	0.5-0.6
G. muris cysts	30-630	1400	7.2-18.5	1.8-2.0

From: Hoff:(28)

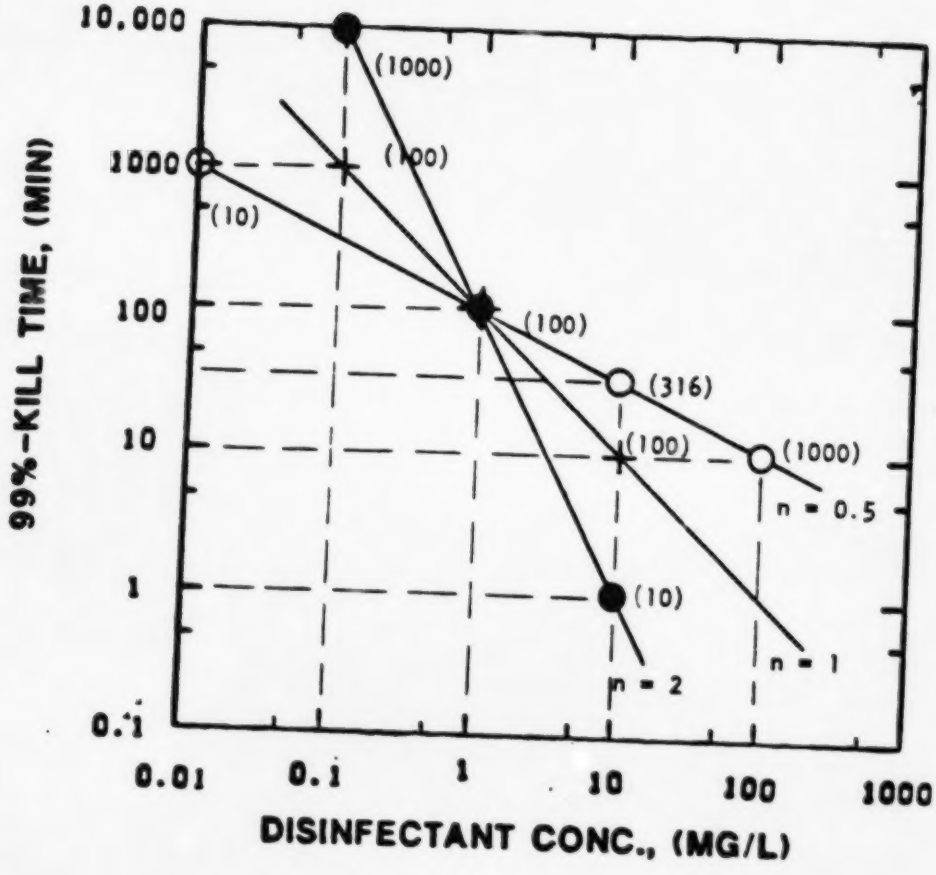

Fig. 1. Effect of n. Value on C·t Values at Different Disinfectant Concentrations (C·t Values in Parentheses)

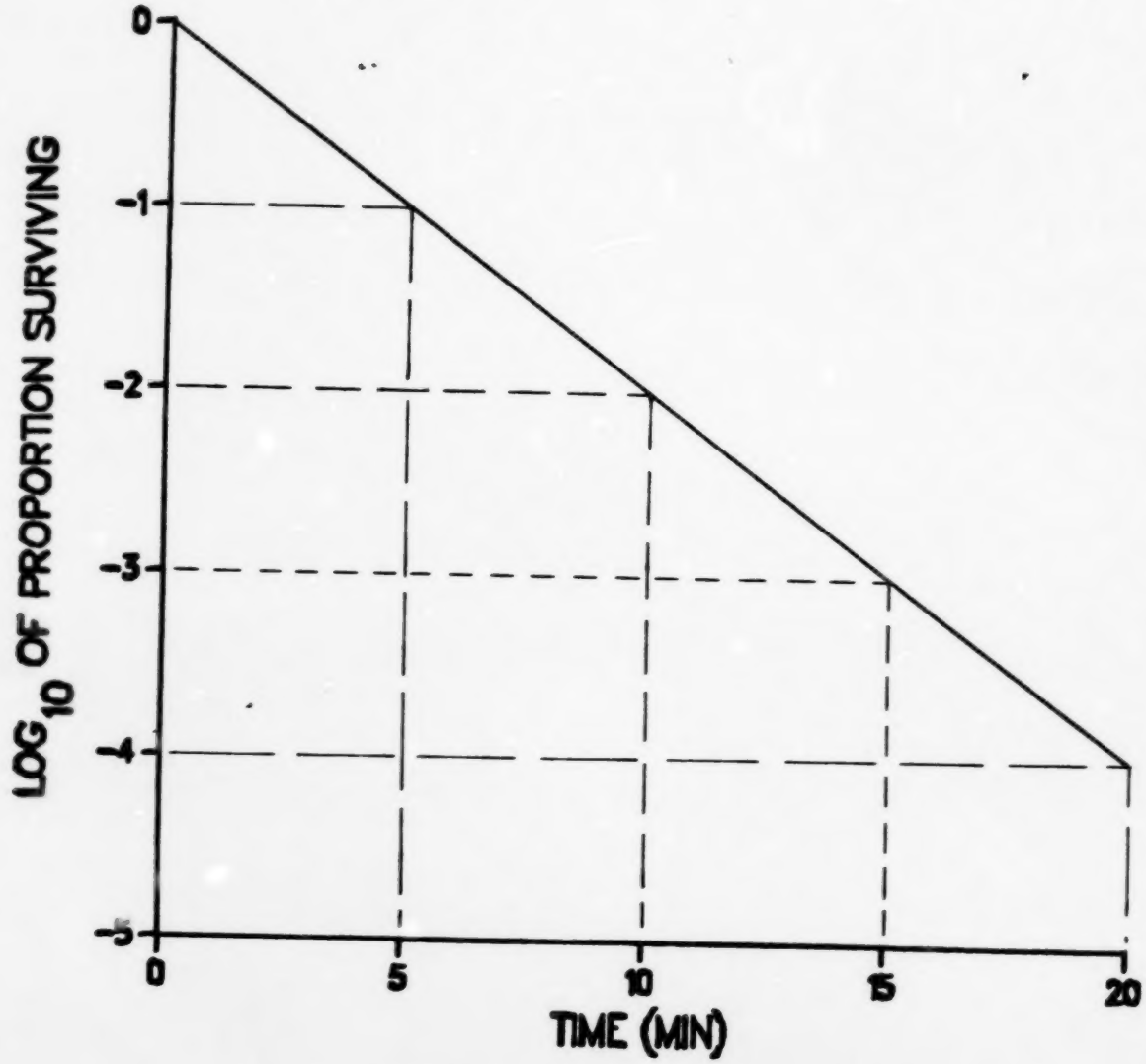

Fig. 2. Inactivation According to Chick's Law

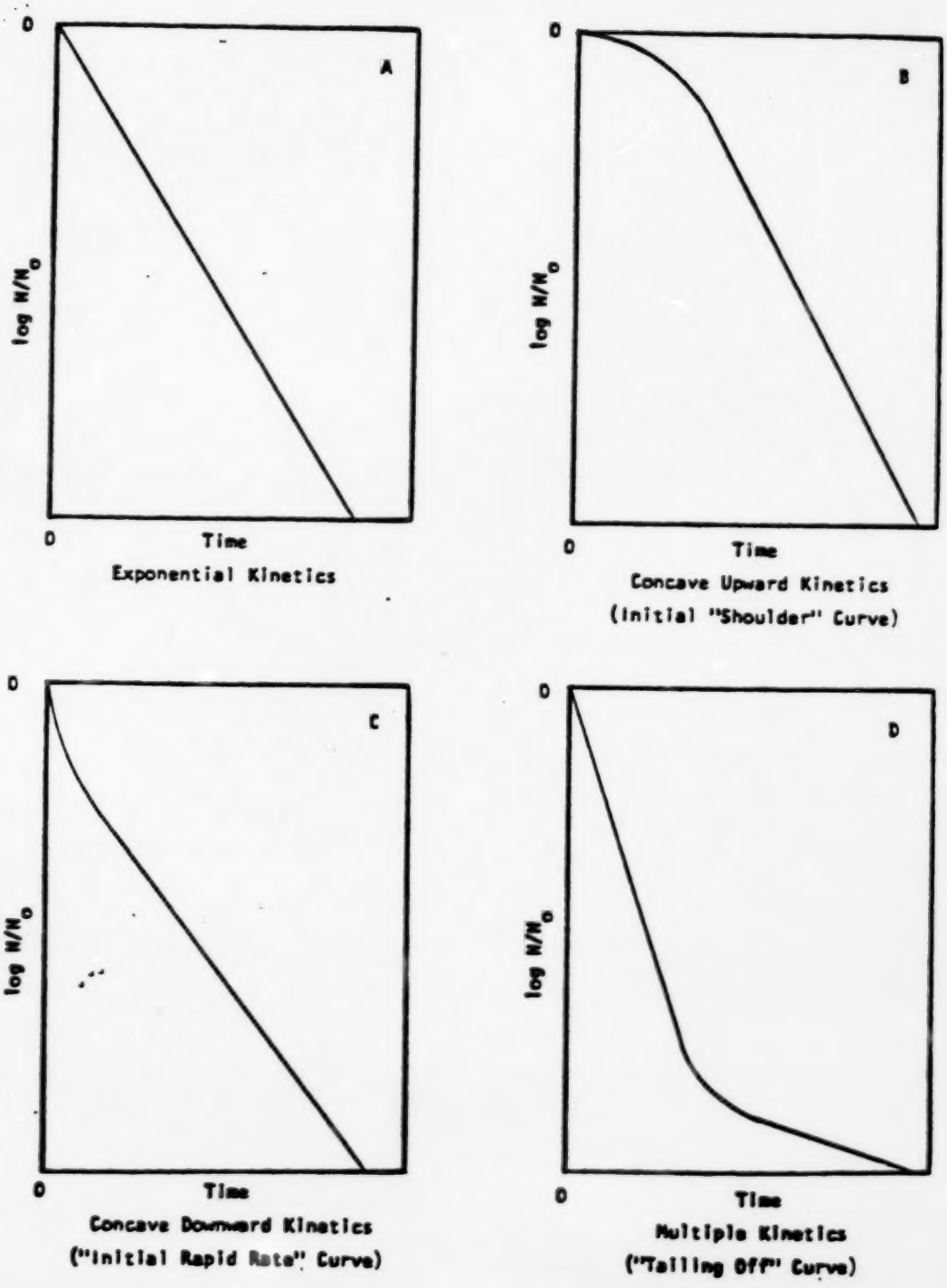

Fig. 3. Typical survival curves for disinfection experiments. After: Prokop and Humphrey (12)